Non-Fermi Liquid Angle Resolved Photoemission Line Shapes of $Li_{0.9}Mo_6O_{17}$

A recent Letter by Xue *et al.* [1] reports observations from angle resolved photoemission spectroscopy (ARPES) on quasi-one-dimensional (quasi-1D) $Li_{0.9}Mo_6O_{17}$ that above its $T_X \approx 24$ K transition a peak dispersing to define its Fermi surface develops Fermi energy (E_F) weight requiring a Fermi liquid (FL) description. This finding contradicts our report [2] of a non-FL line shape in this material. The reasoning in [1] that this new finding was enabled by improved angle resolution is flawed. Rather the data of [1] have fundamental differences from other ARPES results and also band theory, and so the claims of [1] must be held in abeyance. These claims also include the report of an 80 meV gap below T_X, which contradicts the zero gap found in optical spectroscopy [3] and magnetic susceptibility and immensely exceeds the value (0.3 meV) implied by a gap-model interpretation of the resistivity rise below T_X [4].

Improved angle resolution is not relevant for the claimed FL line shape. Because the $\mathbf{k} = \mathbf{k}_F$ line shapes for both the FL and the Tomonaga-Luttinger (TL) (with $\alpha < 1$) models are singular at E_F, the E_F weight for both increases steadily as the angle resolution improves. Indeed, it is well known that one must angle integrate ($\mathbf{k}$ sum) to test for the surprising difference between the Fermi edge of the FL and the E_F power law onset of the TL model. Xue *et al.* sum ARPES data along the quasi-1D Γ-Y direction over $\Delta\mathbf{k} = 0.2$ Å^{-1} and report a Fermi edge, whereas we always find only a power law onset at E_F in angle summed spectra, including our new high resolution spectra shown below. This difference arises because the data are fundamentally different.

Figure 1 shows various Γ-Y data sets for $T > T_X$. Panel (a) shows previously unpublished data taken by us at photon energy $h\nu = 24$ eV on literally the same cleaved surface as for the data of [2]. Panel (b) shows earlier data with $h\nu = 21.2$ eV by Grioni *et al.* [5]. The data sets for the two $h\nu$ values are consistent, both showing bands A through D in good basic agreement with band theory, as labeled. Only bands C, D cross E_F, becoming degenerate before the crossing. Panels (a) and (b) establish the consistency of the data of [2] and [5]. For the special $\mathbf{k}$ path parallel to Γ-Y in [2] both bands C and D are especially strong all the way to E_F. The bands C, D in (a)–(c) along Γ-Y are weaker, but in basic agreement with those in [2] [short dashed lines in (a)], and, importantly, show non-FL line shapes as do the data of [2].

Panel (c) shows $h\nu = 24$ eV data taken at the Wisconsin Synchrotron Radiation Center PGM beam line with an SES-200 Scienta analyzer over a narrow $\mathbf{k}$ range near $\mathbf{k}_F$, where D is already too weak to see. These data have angle and energy resolution comparable to that of Xue *et al.* and fully agree with the data of (a) and (b), apart from generally increased sharpness and increased

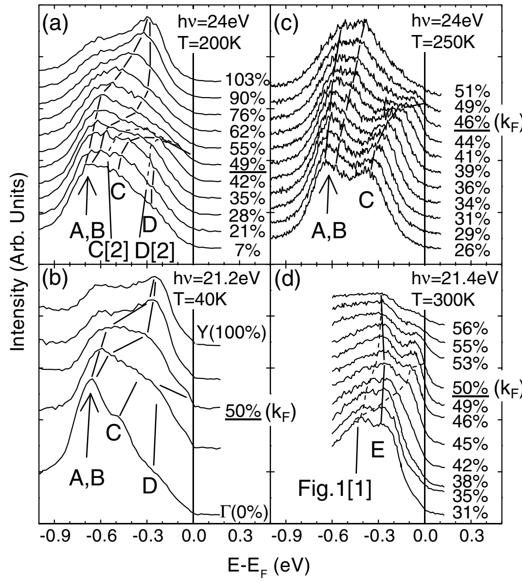

FIG. 1. ARPES data of $Li_{0.9}Mo_6O_{17}$ along $\Gamma(0\%) \rightarrow Y(100\%)$. Energy and angle resolutions are (a) 100 meV, $\pm 1°$, (b) 15 meV, $\pm 1°$ [5], (c) 35 meV, $\leq \pm 0.25°$, and (d) 33 meV, $\pm 0.1°$ [1].

E_F weight for $\mathbf{k}_F$. It is then meaningful to compare the data of (c) directly to the $h\nu = 21.4$ eV data of Xue *et al.* [1], shown in panel (d) with their reported two dispersions in short-dashed lines. Relative to $\mathbf{k}_F$, the two $\mathbf{k}$ ranges are nearly the same. It is obvious by inspection that the two data sets are globally different, for example by the absence in (d) of peaks A, B and by the presence in (d) of a nondispersing feature E which interferes with the presumed C, D line shapes, and has no counterpart in the other data or in band theory.

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